Theory of Ultrafast Optical Manipulation of Electron Spins in Quantum Wells

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Based on a multi-particle-state stimulated Raman adiabatic passage approach, a comprehensive theoretical study of the ultrafast optical manipulation of electron spins in quantum wells is presented. In addition to corroborating experimental findings [Science **292**, 2458 (2001)], we improve the expression for the optical-pulse-induced effective magnetic field, in comparison with the one obtained via the conventional single-particle ac Stark shift. Further study of the effect of hole-spin relaxation reveals that while the coherent optical manipulation of electron spin in undoped quantum wells would deteriorate in the presence of relatively fast hole-spin relaxation, the coherent control in doped systems can be quite robust against decoherence. The implications of the present results on quantum dots will also be discussed.

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I. INTRODUCTION

The electron spin, which has been largely ignored in conventional charge-based electronics, has now become a focus of research due to the emerging field of spintronics [1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12]. It is widely believed that incorporating the spin degree of freedom in conventional charge-based electronics or using electron spin alone as the information carrier will lead to various applications in quantum devices. In particular, various spin-based quantum computer models have been proposed [13, 14, 15, 16, 17, 18, 19, 20].

The development of spintronics largely depends on the ability to control electron spins. One of the most basic controls is coherent spin rotation, which is traditionally implemented by making use of local magnetic fields. However, in order to manipulate the desired coherent spin evolution, the magnetic field pulses should in general be shorter than the electron spin coherence time, which is typically on the order of nanoseconds [21]. Unfortunately, a sub-nanosecond magnetic field source is technologically inaccessible at present. Very recently, several theoretical studies have considered the possibility of manipulating electron spin in solids by *all-optical* means [9, 10, 11]. This approach has significant advantages since the laser field is much more controllable than its magnetic-field counterpart, and tunable femtosecond lasers are now commercially available. In fact, the ultrafast optical manipulation of electron spins in quantum wells has been demonstrated experimentally [3]. The result has been understood in terms of an effective magnetic field arising from the ac Stark shift of the single-electron state [3, 22].

In this paper, we present a many-particle-state theory for the ultrafast optical manipulation of electron spins in quantum wells via the stimulated Raman adiabatic passage (STIRAP) control scheme, in which the effect of Pauli exclusion will be fully taken into account. Furthermore, the consequences of hole-spin relaxation in the valence band will be analyzed in detail, for both an undoped system closely related to the experiment of Ref. 3 and a doped system which has not yet been experimentally investigated. The remainder of this paper is organized as follows. In Sec. II, the concept of effective magnetic field, which is crucial to the optical manipulation of electron spin, is introduced first by considering the ac Stark shift of single-particle state. A many-particle STIRAP approach to coherent spin control is then presented, together with numerical and perturbative results. The effect of hole-spin relaxation is studied in Sec. III. Doped systems will be considered in Sec. IV, where both the control of STIRAP manipulation and the effect of hole-spin relaxation are studied in comparison with their undoped counterparts considered earlier. Finally, concluding remarks are presented in Sec. V.

II. COHERENT MANIPULATION

ac Stark Effect and the Description of Effective Magnetic Field.— The basic idea of an optical approach to manipulating electron spin is to make use of an off-resonance laser pulse to induce ac Stark shifts, which are in turn equivalent to an effective magnetic field [23, 24, 25]. This idea has recently been employed to manipulate electron spin in undoped semiconductor quantum wells [3]. Initially, an electron is pumped from the valence band to the conduction band by a resonant laser pulse along the z-direction and with σ^+-polarization. The resultant state diagram is shown in Fig. 1(a), where all the states are in the "z"-representation [26]. In the absence of magnetic field, the lowest conduction-band (CB) level is two-fold degenerate, denoted by spin states $|\pm 1/2\rangle_c$; and the valence-band (VB) states are denoted by $|\pm 3/2\rangle_v$ and $|\pm 1/2\rangle_v$. Here, we have taken into account the splitting of the valence band-edge states from

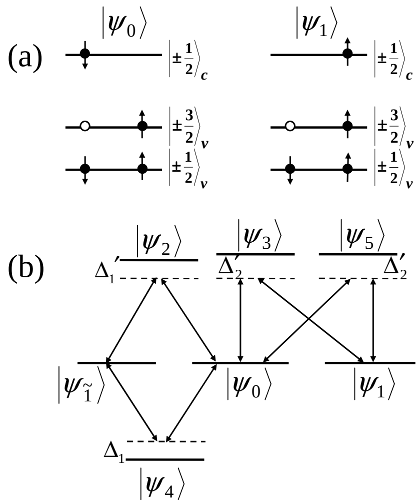

FIG. 1: Diagram of band-edge states of undoped quantum well after optically exciting an electron from the valence to the conduction band. (a) Two degenerate state configurations with different electron spin in the conduction band, between which quantum coherent oscillation is to be performed. (b) Intermediate states which mediate transition between $|\psi_0\rangle$ and $|\psi_1\rangle$ during the optical manipulation.

the original four-fold degenerate states into two two-fold degenerate states, due to the breakdown of the symmetry of the quantum well. Further consideration of the exciton effect leads to the shifted electron-hole transition energies, $E_{X_1} = (E_c - E_{v_1}) - U_{X_1}$ and $E_{X_2} = (E_c - E_{v_2}) - U_{X_2}$. Here E_c, E_{v_1} and E_{v_2} denote the energies of $|\pm 1/2\rangle_c$, $|\pm 3/2\rangle_v$ and $|\pm 1/2\rangle_v$, respectively, and U_{X_1} and U_{X_2} are the Coulomb interaction (attraction) energies of the corresponding electron-hole pairs. The discrete-level model of Fig. 1(a) for a quantum well is based on two considerations: (i) the initial preparation of the electron-hole pair is accomplished by the resonant excitation between the band-edge states; (ii) the subsequent off-resonant optical manipulation involves only the band-edge states because of the negligibly small momenta of the photons causing only vertical (and also virtual) transitions. Therefore, states in the diagram shown in Fig. 1(a) should be understood as the band-edge states.

To manipulate the electron spin in the conduction band, let us apply a below-band-gap (negative-detuning) laser pulse of the same σ^+-polarization but along the x-direction. This laser induces $|-1/2\rangle_{c,x} \leftrightarrow |-3/2\rangle_{v,x}$ and $|1/2\rangle_{c,x} \leftrightarrow |-1/2\rangle_{v,x}$ transition couplings between the specified states in the "x"-representation. Other transitions are forbidden due to the selection rule. It is well known that this kind of off-resonance coupling will cause ac Stark shifts of the relevant energy levels. In particular, from second-order perturbation theory the energy shifts of the CB states $|-1/2\rangle_{c,x}$ and $|1/2\rangle_{c,x}$ can be estimated as [9]:

$$\Delta E^{c,x}_{-1/2} = |V_{-1/2,-3/2}|^2/\Delta_1, \tag{1a}$$

$$\Delta E^{c,x}_{1/2} = |V_{1/2,-1/2}|^2/\Delta_2. \tag{1b}$$

Here, $\Delta_1 = E_{X_1} - \hbar\omega_p$ and $\Delta_2 = E_{X_2} - \hbar\omega_p$ are the detunings of the photon energy ($\hbar\omega_p$) with respect to the excitation energies from the heavy and light valence band states, respectively, to the conduction band state. $V_{-1/2,-3/2}$ and $V_{1/2,-1/2}$ are the coupling matrix elements of the laser with the electron-hole pair states. Explicitly, from energy-band theory [27], the spatial wave-functions of the considered states are composed of the Bloch functions $|S\rangle$, $|X\rangle$, $|Y\rangle$ and $|Z\rangle$. Denoting $\Omega \equiv eE_0\langle S|x|X\rangle = eE_0\langle S|y|Y\rangle = eE_0\langle S|z|Z\rangle$, the coupling matrix elements read

$$\begin{aligned} V_{-1/2,-3/2} &= eE_{0c,x}\langle -1/2|\vec{r}\cdot\vec{\epsilon}|-3/2\rangle_{v,x} \\ &= -ieE_0\langle S|y+iz|Y-iZ\rangle/\sqrt{2} = -i\sqrt{2}\Omega, \end{aligned} \tag{2a}$$

$$\begin{aligned} V_{1/2,-1/2} &= eE_{0c,x}\langle 1/2|\vec{r}\cdot\vec{\epsilon}|-1/2\rangle_{v,x} \\ &= ieE_0\langle S|y+iz|Y-iZ\rangle/\sqrt{6} = i\sqrt{2/3}\Omega. \end{aligned} \tag{2b}$$

Here the equation $\vec{r}\cdot\vec{\epsilon} = y + iz$ stems from the σ^+-polarization and x-direction propagation of the laser pulse.

In practice, the ac Stark shifts can be conveniently described as an effective magnetic field B_{eff} along the manipulating laser direction. In terms of the CB level splitting $\delta_{CB} = \Delta E^{c,x}_{-1/2} - \Delta E^{c,x}_{1/2}$, the effective magnetic field reads

$$B_{eff} = \delta_{CB}/(g_e\mu_B). \tag{3}$$

Here g_e is the Landé-g factor and μ_B is the Bohr magneton. In addition, the duration of this effective magnetic field is equivalent to the duration of the laser-pulse, which can be as short as femtoseconds. This effective magnetic field description can be further elaborated as follows. The initially created CB electron spin state $|1/2\rangle_c$ is an eigenstate of the Pauli operator σ_z. It can be expressed in terms of the eigenstates of σ_x as $|\Psi(t=0)\rangle = |1/2\rangle_c = \frac{1}{\sqrt{2}}(|1/2\rangle_{c,x} + |-1/2\rangle_{c,x})$. Upon switching on the off-resonance laser pulse along the x-direction, the ac Stark shifts $\Delta E^{c,x}_{\pm 1/2}$ would result in a relative phase difference $\phi = (\Delta E^{c,x}_{1/2} - \Delta E^{c,x}_{-1/2})t$, between the states $|1/2\rangle_{c,x}$ and $|-1/2\rangle_{c,x}$. Recasting $|\Psi(t)\rangle$ in the σ_z-eigenstate representation leads to $|\Psi(t)\rangle = \cos\frac{\phi}{2}|1/2\rangle_c + i\sin\frac{\phi}{2}|-1/2\rangle_c$. Clearly, the off-resonant laser pulse along the x-direction plays the role of an effective magnetic field pulse with magnitude determined by Eq. (3).

Many-Particle STIRAP Approach.— In the following we present an improved effective magnetic field description based on a many-particle state STIRAP approach. This approach is a direct generalization of the single-particle STIRAP in quantum optics [28] that takes into account the multi-electron occupation of the valence band. The effective coupling between $|1/2\rangle_c$ and $|-1/2\rangle_c$

will then be established via a number of intermediate many-electron states as depicted in Fig. 1(b).

For convenience, we introduce Fock's particle-number representation to denote the multi-electron state. For instance, we denote $|\psi_0\rangle \equiv |1,0;0,1,1,1\rangle$, and $|\psi_1\rangle \equiv |0,1;0,1,1,1\rangle$, where "1" ("0") stands for the occupation (vacancy) of the individual single particle states, which correspond to, respectively, the CB states $|\mp 1/2\rangle_c$, and the VB states $|\mp 3/2\rangle_v$ (heavy holes) and $|\mp 1/2\rangle_v$ (light holes). Under the action of the off-resonance laser pulse described previously, the CB states will be virtually coupled to the VB states. As a consequence, a number of intermediate virtual states establish an effective interaction between $|\psi_0\rangle$ and $|\psi_1\rangle$. Depending on the polarization of the laser pulse, selection rules only allow the following intermediate states [27, 30]: $|\psi_2\rangle \equiv |0,0;1,1,1,1\rangle$, $|\psi_3\rangle \equiv |1,1;0,1,1,0\rangle$, $|\psi_4\rangle \equiv |1,1;0,0,1,1\rangle$, and $|\psi_5\rangle \equiv |1,1;0,1,0,1\rangle$. Also, based on the selection rule, a possible state, $|\psi_{\tilde{1}}\rangle \equiv |0,1;1,0,1,1\rangle$, may be correlated with the initial state $|\psi_0\rangle$. All these relevant multi-electron states are shown in Fig. 1(b). In the absence of spin relaxation, the subspace spanned by these basis states is complete for the evolution of the system. We refer to this subspace as *coherent subspace*, and denote it by $\mathbf{M}^{coh} = \{|\psi_i\rangle, i = 0,1,\tilde{1},2,3,4,5\}$. In $\mathbf{M}^{coh}$ the driving Hamiltonian (in the interaction picture) reads

$$H = \begin{pmatrix} 0 & 0 & 0 & \Omega_{20}^* & \Omega_{30}^* & \Omega_{40}^* & \Omega_{50}^* \\ 0 & 0 & 0 & 0 & \Omega_{31}^* & 0 & \Omega_{51}^* \\ 0 & 0 & 0 & \Omega_{2\tilde{1}}^* & 0 & \Omega_{4\tilde{1}}^* & 0 \\ \Omega_{20} & 0 & \Omega_{2\tilde{1}} & -\Delta_1 & 0 & 0 & 0 \\ \Omega_{30} & \Omega_{31} & 0 & 0 & \Delta_2' & 0 & 0 \\ \Omega_{40} & 0 & \Omega_{4\tilde{1}} & 0 & 0 & \Delta_1' & 0 \\ \Omega_{50} & \Omega_{51} & 0 & 0 & 0 & 0 & \Delta_2' \end{pmatrix}. \tag{4}$$

Here $\Delta_1' = \Delta_1 + U_{XX}$, and $\Delta_2' = \Delta_2 + U_{XX}$, where U_{XX} is the Coulomb repulsive energy of two excitons that appear in the intermediate virtual states. In Eq. (4), $\Omega_{ij} = eE_0\langle\psi_i|\vec{r}\cdot\vec{\epsilon}|\psi_j\rangle$ describes the laser-induced coupling between the conduction and valence band states. Similar to the calculation in Eq. (2), Ω_{ij} can be straightforwardly determined by using

$$eE_0\langle S|y+iz|X+iY\rangle = i\Omega, \tag{5a}$$

$$eE_0\langle S|y+iz|X-iY\rangle = -i\Omega, \tag{5b}$$

$$eE_0\langle S|y+iz|Z\rangle = i\Omega. \tag{5c}$$

Note that here all the states are expressed in the "z"-representation, while the states in Eq. (2) are in the "x"-representation. Explicitly, we obtain $\Omega_{20} = \Omega_{4\tilde{1}} = -\Omega_{40} = -\Omega_{2\tilde{1}} = -\Omega/\sqrt{2}$, $\Omega_{50} = \Omega_{31} = \Omega/\sqrt{6}$, $\Omega_{30} = \Omega_{51} = -\sqrt{2/3}\Omega$.

In analogy to the (three-state) Λ atomic system in quantum optics [28], the problem under study can now be solved via a many-particle multi-state STIRAP approach. Before giving a numerical demonstration, let us consider an effective many-particle two-state Hamiltonian approach to the description of the STIRAP coupling

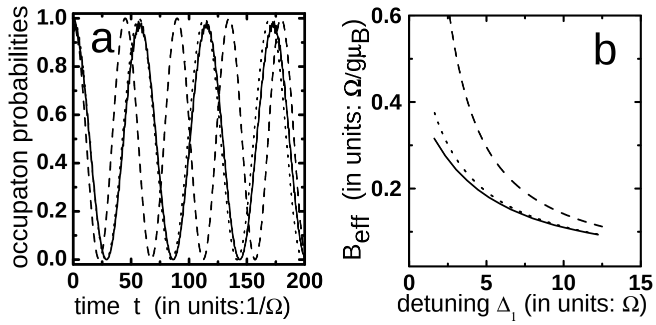

FIG. 2: Coherent manipulation of electron spin in the absence of hole spin relaxation in the valence band. (a) Laser-induced Rabi oscillation between the spin-up and spin-down states, where the results depicted by the solid, dotted and dashed curves are respectively from three approaches: the direct numerical calculation with the coherent subspace Hamiltonian of Eq. (4), the effective two-state multi-particle STIRAP Hamiltonian of Eq. (6) or (7), and the analysis based on the single-particle-state picture of Eq. (3). (b) The corresponding effective magnetic fields extracted from these three methods. Here Δ_1 denotes the detuning.

between $|\psi_0\rangle$ and $|\psi_1\rangle$. Notice that $|\psi_0\rangle$ also virtually couples to $|\psi_{\tilde{1}}\rangle$ through two paths [cf. Fig. 1(b)] via the intermediate states $|\psi_2\rangle$ and $|\psi_4\rangle$, respectively. However, their opposite excitation configurations lead to cancellation of these two paths due to destructive interference, provided that the magnitude of the detuning is much larger than the exciton interaction energy U_{XX}. The above analysis will be verified numerically soon. Therefore, we may only need to consider the two paths connecting $|\psi_0\rangle$ and $|\psi_1\rangle$ via $|\psi_3\rangle$ and $|\psi_5\rangle$. Adiabatically eliminating the intermediate states leads to the effective two-state multi-particle STIRAP Hamiltonian [28]

$$H_{eff} = -\tilde{\Omega}(|\psi_0\rangle\langle\psi_1| + |\psi_1\rangle\langle\psi_0|)/2, \tag{6a}$$

with the Rabi frequency

$$\tilde{\Omega}/2 = |\Omega_{31}\Omega_{30} + \Omega_{51}\Omega_{50}|/\Delta_2'. \tag{6b}$$

By identifying the Rabi frequency with the Larmor frequency of spin precession around a magnetic field, the *effective magnetic field* can be defined as

$$\tilde{B}_{eff} = \tilde{\Omega}/(g_e\mu_B). \tag{7}$$

Remarkably, the following numerical calculation will demonstrate that this is a considerable improvement over Eq. (3), as a result of taking into account the multi-electron occupation in the valence band.

Numerical Results.— In accordance with the experiment of Ref. 3, where the spin rotation period under optical manipulation is on a timescale of picoseconds, we choose the following parameter values in our numerical study: the Rabi frequency $\Omega = 4$ meV, the detuning parameters $\Delta_1 = 10\,\Omega$ and $\Delta_2 = 11.5\,\Omega$, and the exciton Coulomb interaction energy $U_{XX} = 0.5\,\Omega$. Figure 2(a)

depicts the laser induced Rabi oscillations between the spin-up and spin-down states of the CB electron. We present results from three approaches: the direct numerical calculation with the coherent subspace Hamiltonian of Eq. (4), the effective two-state multi-particle STIRAP Hamiltonian of Eq. (6) or (7), and the analysis based on the single-particle-state picture of Eq. (3). We find that the effective two-state multi-particle STIRAP Hamiltonian approach gives a result almost identical with the exact numerical calculation, while the result based on the single-particle ac Stark shift differs considerably. As we have already analyzed, the Rabi oscillation of spin can be equivalently described as a the Larmor precession around an effective magnetic field. In Fig. 2(b) we show the effective magnetic field complementary to the Rabi oscillation in Fig. 2(a). In the adiabatic regime (i.e. the large detuning case), the multi-particle-state-based effective magnetic field given by Eq. (7) can precisely describe the CB electron spin rotations. In contrast, the single-particle-state-based counterpart given by Eq. (3) is inaccurate and will cause considerable errors in practice.

So far, we have only considered the rotation of the conduction electron spin. Now we briefly discuss the possible effects of hole-spin rotation, which has been typically ignored in the analysis of Faraday rotation experiments [3, 4] because of its rapid relaxation with respect to the conduction electron spin. In general, the hole-spin rotation should also affect the Faraday rotation signals in the coherent (or short-time) regime. Interestingly, it can be shown that for the specific optical excitation-manipulation setup studied in this work, the off-resonant laser along the x-direction rotates only the conduction electron spin but does not affect the hole spin at all. Of course, both the electron and hole spins would be rotated if the manipulating laser propagates along a different direction. Probing both the electron and hole spin rotations should be an interesting subject of Faraday rotation experiments in general.

III. EFFECT OF HOLE SPIN RELAXATION

In reality, both the CB electron spin and VB hole spin will suffer environment-induced scattering, and have finite decoherence times. In Ref. 11, electron-hole recombination was considered the dominant source of decoherence for quantum dots. For the quantum well studied in this work, however, the dominant source of decoherence should be the VB hole-spin relaxation. It typically occurs in a matter of picoseconds [3, 4, 31, 32], much shorter than the typical timescale of nanoseconds for electron-hole recombination and CB electron spin relaxation [3, 4, 21].

The hole-spin relaxation involves the following incoherent VB state jumps: $|3/2\rangle_v \leftrightarrow |-3/2\rangle_v$, $|1/2\rangle_v \leftrightarrow |-1/2\rangle_v$, and $|\pm 3/2\rangle_v \leftrightarrow |\pm 1/2\rangle_v$. These processes can be described with the spin-jump operators $S_1 = |-3/2\rangle_v\langle 3/2|$, $S_2 = |-1/2\rangle_v\langle 1/2|$, and $S_{3,4,5,6} = |\pm 1/2\rangle_v\langle \pm 3/2|$, respectively, together with their Hermitian conjugates. In particular, we employ the well-known Lindblad master equation to address the hole spin relaxation effect, which reads [28, 29]

$$\dot{\rho} = -i[H,\rho] - \sum_j \gamma_j D[S_j] - \sum_j \tilde{\gamma}_j D[S_j^\dagger]. \qquad (8)$$

The Lindblad superoperators are $D[S_j] = \frac{1}{2}\{S_j^\dagger S_j, \rho\} - S_j \rho S_j^\dagger$ and $D[S_j^\dagger] = \frac{1}{2}\{S_j S_j^\dagger, \rho\} - S_j^\dagger \rho S_j$, associated with the hole-spin relaxation strengths of γ_j and $\tilde{\gamma}_j$, respectively. In the following numerical studies we assume an identical strength of $\gamma_j = \tilde{\gamma}_j \equiv \gamma_0 = 0.1\,\Omega$. This choice assumes a similar timescale for the hole-spin relaxation and the laser-induced Rabi oscillation period of the electron spin. It thus allows us to demonstrate in a transparent manner the effect of hole-spin relaxation on optical manipulation. Also, this choice is qualitatively consistent with the experiment in Ref. 3.

In the presence of hole-spin relaxation, the restricted subspace $\mathbf{M}^{coh}$ constructed for the coherent evolution is no longer complete. More specifically, the relaxation operators S_j and $S_j^\dagger$ ($j = 1, \ldots 6$) will cause a leakage of state from the subspace $\mathbf{M}^{coh}$. By applying the relaxation operators to the basis states of subspace $\mathbf{M}^{coh}$, eight new states outside the subspace $\mathbf{M}^{coh}$ appear. We now denote the expanded Hilbert space by $\mathbf{M}^{inc} = \{|\psi_i\rangle, i = 0, 1, \tilde{1}, 2, 3, 4, 5; |\tilde{\psi}_j\rangle, j = 1, 2, ...8\}$, which is complete for the evolution of Eq. (8). Using the Lindblad equation (8), in the following we study the hole-spin relaxation effect for two possible situations.

Instant manipulation.— In this case the spin manipulation is performed right after the generation of the

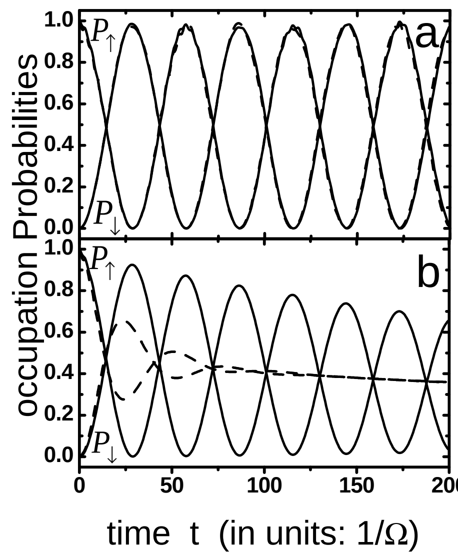

FIG. 3: Hole spin relaxation effect on the manipulation of the conduction-band electron spin, in the absence (a) and presence [(b) and (c)] of pre-relaxation of the hole spin, respectively.

electron-hole pair. Figure 3(a) shows the effect of hole-spin relaxation on the Rabi oscillation of CB electron spin, where we have classified the spin-up and spin-down states according to the CB electron spin, i.e., summing the probabilities of states with the same CB spin state, regardless of the hole spin states of the valence band. The result in Fig. 3(a) shows that the hole-spin relaxation will significantly influence the optical manipulation of the CB electron spin, degrading it out from the desired coherent subspace (and into the incoherent regime). Also, differing from the conventional decoherence effect on *real* two-state Rabi oscillation, besides the decay of the oscillation amplitudes in Fig. 3(a), the sum of the probabilities of spin-up and spin-down states eventually deviates from unity. This deviation is caused by the relaxation-induced real occupation of the intermediate states, which are ruled out from the CB electron spin-up and spin-down states. In the above calculation, we have adopted parameters such that the Rabi oscillation of the CB spin has a period similar to the hole-spin relaxation time. By increasing the laser-system coupling strength, more "coherent" oscillations can be observed. Nevertheless, the present result implies that coherent optical manipulation in the *undoped* quantum well is limited by the hole-spin relaxation, which is typically on a timescale of picoseconds.

Manipulation in the presence of hole-spin pre-relaxation.— In practice (e.g. in the experiment of Ref. 3), the manipulating laser pulse may be switched on with a certain delay time after the resonant excitation pulse that prepared the initially coherent electron-hole pair. It is thus meaningful to show how the *pre-relaxation* of the hole spin affects the subsequent manipulation of the electron spin. Before the manipulating laser is applied, the fast hole-spin relaxation may have led the valence band to a mixed hole state, while the CB electron spin that has not yet coupled to the VB state remains in the initially prepared pure state. As the manipulating laser field is applied, Rabi coupling between the CB pure state and the VB mixed state is established, and relaxations among several involved multi-particle states occur. The optical manipulation in the presence of the above pre-relaxation can be described by using the time-dependent Hamiltonian $H = H_0 + \Theta(t-\tau)H_{int}(t)$ for the same Lindblad master equation (8). Here H_0 is the field-free system Hamiltonian, while $\Theta(t-\tau)H_{int}(t)$, with $\Theta(t-\tau)$ being the step-function, describes the system's interaction with the time τ-delayed manipulating laser field.

Shown in Fig. 3(b) and (c) are the resulting Rabi oscillations of the CB electron spin evaluated with two delay time values . Compared to the instant manipulation counterpart in Fig. 3(a), the results shown here indicate that the pre-relaxation of the VB hole spin does not significantly affect the manipulation of the CB electron spin. Intuitively, since the VB had been in a mixed state, one would not expect any "coherent" signal to appear in response to the optical manipulation, but some coherent signal can be seen in the incoherent regime of Fig. 3(a).

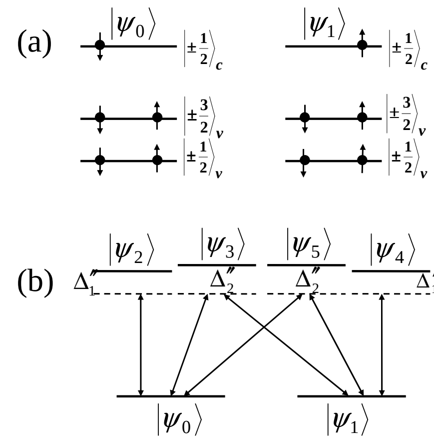

FIG. 4: Diagram of band-edge states of a doped quantum well. (a) Two degenerate state configurations with different electron spin in the conduction band, between which quantum coherent rotation is to be performed. (b) Intermediate states which mediate transition between $|\psi_0\rangle$ and $|\psi_1\rangle$ during the optical manipulation. Here the detunings Δ_1'' and Δ_2'' differ in a certain sense from their counterparts Δ_1' and Δ_2' in the undoped quantum well, i.e., $\Delta_1'' = \Delta_1 + U_{eX}$, and $\Delta_2'' = \Delta_2 + U_{eX}$, with U_{eX} the Coulomb interaction energy of the doped electron and the (virtually) generated exciton.

This counterintuitive result may be understood by noting that in Fig. 3(a) the stationary mixed state is achieved in the presence of the laser pulse. In that completely mixed state, the CB electron spin jumps incoherently between spin-up and spin-down states, and the hole state is not at all the pre-relaxed valence band state achieved at the end of the first stage in Fig. 3(b) and (c). Thus, the laser pulse can still drive coherent oscillation of the CB electron spin as in Fig. 3(a). As a matter of fact, the analysis presented here corresponds to the experiment in Ref. 3, where the tipping laser pulse acted on a pre-relaxed state. The experimental result showed that the electron spin can still be manipulated via the intermediate mixed valence band state by the laser pulse, which is consistent with our present understanding.

IV. DOPED SEMICONDUCTORS

So far our discussion has focused on the undoped semiconductor quantum well, where the excitation pump field creates an electron in the CB while leaving a hole in the VB. In this section we shall compare it with the doped quantum well, in which an excess electron is injected into the conduction band, and the valence band remains fully occupied. Shown in Fig. 4(a) is the level diagram of a doped semiconductor quantum well. Compared with the undoped system in Fig. 1(a), no hole exists in the valence band of the doped system. However, an electron in the VB can be virtually excited to the CB during the manipulation. This process is described by a number of interme-

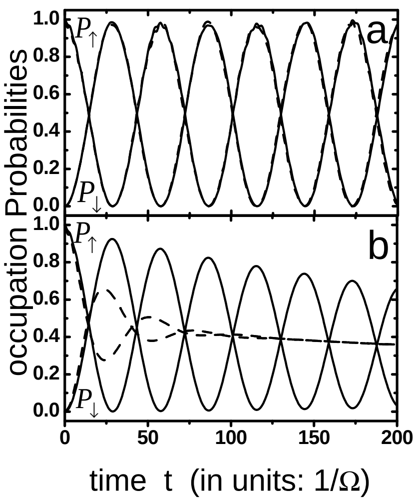

FIG. 5: Comparison of optical manipulation of the conduction-band electron spin in doped and undoped quantum wells, in the (a) absence and (b) presence of hole-spin relaxation. The solid and dashed curves correspond to, respectively, results of the doped and undoped quantum wells.

diate states shown in Fig. 4(b). Using the same notation as for the undoped system, we denote the initial state as $|\psi_0\rangle \equiv |1,0;1,1,1,1\rangle$, the (conduction band) spin-flipped state as $|\psi_1\rangle \equiv |0,1;1,1,1,1\rangle$, and the relevant virtually excited intermediate states as $|\psi_2\rangle \equiv |1,1;1,0,1,1\rangle$, $|\psi_3\rangle \equiv |1,1;1,1,1,0\rangle$, $|\psi_4\rangle \equiv |1,1;1,1,0,1\rangle$, and $|\psi_5\rangle \equiv |1,1;0,1,1,1\rangle$. With the identification of these intermediate states, the same analysis as for the undoped system can be carried out for both the coherent manipulation and the hole-spin relaxation effect. In the following calculation, we assume $U_{XX} \simeq U_{eX}$. Any possible difference between them would be minor in comparison with the large detuning energy, resulting only in a negligibly small change of the Rabi oscillation frequency. As shown in Fig. 5(a), the doped and undoped systems have almost identical responses to optical manipulation in the absence of hole-spin relaxation. However, in the presence of hole-spin relaxation, the doped and undoped systems behave very differently. As shown in Fig. 5(b), while coherent manipulation in undoped semiconductor quantum wells is sensitively limited by the hole-spin relaxation that virtually exists in the intermediate states, optical manipulation of the doped system is much more robust. This result indicates that satisfactory coherent manipulation should be achievable in doped systems.

V. CONCLUDING REMARKS

To summarize, we have presented a theoretical study of the optical manipulation of electron spins in semiconductor quantum wells. Our results provide not only a qualitative account for the experimental findings in Ref. 3, but also a considerably improved effective magnetic field description based on the present many-particle-state STIRAP approach. Moreover, we have also analyzed the effect of hole-spin relaxation, and showed that while coherent optical control in undoped quantum wells deteriorates significantly in the presence of relatively fast hole-spin relaxation, coherent control in doped systems can be quite robust against decoherence.

We would also like to mention that similar analysis based on the present formalism can be straightforwardly applied to the optical manipulation of spins in quantum dots, a topic of great interest in recent years [6, 7, 8, 10, 11, 13, 14, 15, 16, 17, 18, 19, 20]. In a quantum dot, the energy levels are discrete, and thus long spin relaxation time are anticipated. In fact it has been shown experimentally that in quantum dots both the electron and hole spins are almost frozen within the electron-hole recombination timescale [33], which is on the order of nanoseconds or longer. Recent calculations predicted that for a typical quantum dot the conduction electron spin relaxation time is about $10^{-6} \sim 10^{-4}$ seconds [34], and the hole-spin relaxation time is of order 10^{-8} seconds or longer [35]. These results suggest that the coherent optical manipulation of electron spins discussed in this work can readily be carried out in quantum dots. Further, based on the result presented in Sec. IV, almost identical control quality within the coherent time scale is expected for either doped or undoped systems. The latter is in general more experimentally accessible. With optical means, the coherent conduction electron spin rotation can be performed thousands of times before the electron-hole recombination.

Finally, we remark that the present work has focused on quantum wells. Therefore, the applicability of the discrete-level model should be restricted to the optical manipulation of band-edge states. Also, in our treatment the electron-hole Coulomb interaction (exciton effect) is only accounted for in terms of some phenomenological binding-energy parameters. Although such treatment is typical for the optical properties of quantum wells, a more sophisticated treatment based on certain advanced techniques of many-body theory might be desirable in future work. Also, in the study of the hole-spin relaxation effect, the relaxation strengths have been put only by hand. To make the study more realistic, it would be desirable to present calculations of the scattering rates between various states based on the microscopic mechanisms.

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Fig.3 by Jin and Li